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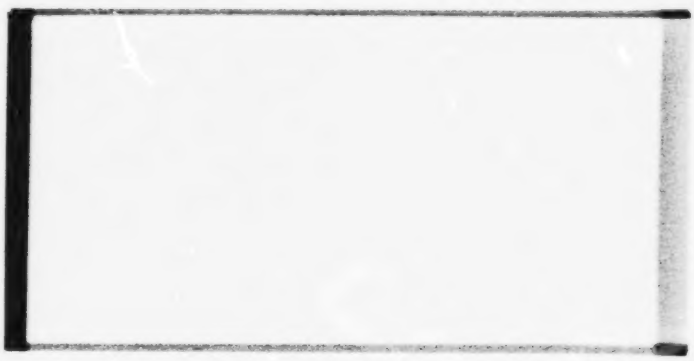
REVIEW OF GAS TREATMENT SYSTEMS FOR NUCLEAR FACILITIES

Report No 82-139-K

C.H. Cheh
Environmental & Inorganic Section
Chemical Research Department



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Reviewed by: J. Chadha
Area Coordinator

ABSTRACT

Gaseous radionuclide emission control technology for normal station operation, for post-LOCA containment clean-up in nuclear generating stations and for fuel reprocessing plants are reviewed and compared. Gas treatment or ventilation systems required to control the emissions of most of the radionuclides of concern from stations are relatively simple and are commercially available. The main emphasis of R&D should be on improving the operation and efficiency of these systems. Tracer gas techniques can be applied to identify the radionuclide release pathways and develop effective control schemes and should therefore be further tested. Studies on the Tritium Removal System and control technologies for carbon-14 and noble gases should also be carried out. Fuel reprocessing off-gas treatment systems are mostly in the developmental stage except for iodine and particulates. Most of these processes have not been tested in integrated off-gas systems where interference by other species present may occur. Hence R&D on processes for individual radionuclides and integrated treatment systems are required.

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740631-141-461	837.81	December 31, 1982	82-139-K

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EXECUTIVE SUMMARY

REVIEW OF GAS TREATMENT SYSTEMS FOR NUCLEAR FACILITIES

C.H. Cheh
Environmental & Inorganic Research Section
Chemical Research

AECL's off-gas development program is mainly on systems for reactor wastes and safety as well as fuel/waste processing facilities whereas Ontario Hydro's interests are more oriented to emission control options for nuclear generating stations. Because of the limited resources in the two organizations, it was decided to attempt to determine what common R&D areas there are in terms of airborne radionuclide control for the three types of operations, namely nuclear station during normal operation, post-LOCA containment clean-up and fuel reprocessing plants (FRP). These data would then be used as the basis for coordinating AECL and OH's efforts in the development of off-gas treatment systems.

In this report, the designs of gas treatment systems in Pickering 'A' and 'B', Bruce 'A' and 'B' and Darlington are summarized. The status of off-gas control technology for FRP is also reviewed and compared with systems in stations. Gas treatment or ventilation systems required to control the emissions of most of the radionuclides of concern from stations are relatively simple and are commercially available. The main emphasis of R&D should be on improving the operation and efficiency of these systems. Tracer gas techniques can be applied to identify the radionuclide release pathways and develop effective control schemes and should therefore be further tested. Studies on the Tritium Removal System and control technologies for carbon-14 and noble gases should also be carried out. FRP off-gas treatment systems are mostly in the developmental stage except for iodine and particulates. Most of these processes have not been tested in integrated off-gas systems where interference by other species present may occur. Hence R&D on processes for individual radionuclides and integrated treatment systems are required.

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To Mr. H.N. Isaac
Manager
Nuclear Materials Management Department

REVIEW OF GAS TREATMENT SYSTEMS FOR NUCLEAR FACILITIES

1.0 INTRODUCTION

Control technologies for gaseous radionuclide emissions from nuclear generating stations (NGS) during normal station operation and post-loss-of-coolant-accident (post-LOCA) containment clean-up may be, in some cases, applicable to emission control for irradiated fuel reprocessing plants (FRP). The main objective of this report is to provide a basis for establishing a coordinated effort between AECL and Ontario Hydro on off-gas treatment system development by comparing gas treatment systems and identifying R&D areas for the three cases.

In this report, the designs of gas treatment systems in Pickering 'A' and 'B', Bruce 'A' and 'B' and Darlington are summarized. The status of off-gas control technology for FRP is also summarized based on an up-date of the review carried out by Jackson/1/. The gas treatment systems for normal station operation, post-LOCA containment clean-up and FRP are then compared and R&D areas identified.

2.0 SYSTEM DESCRIPTION

Gas treatment systems for normal station operation, post-LOCA containment clean-up and fuel reprocessing plants are described in Appendix A, B and C respectively. The design of the systems at Pickering 'A' and 'B', Bruce 'A' and 'B' and Darlington are reviewed/2,3,4,5,6/ and a brief description of the gas treatment systems for these three cases is given below.

2.1 Normal Station Operation

There are three major gas treatment systems that reduce gaseous radioactive emissions during normal station operation. These are the Off-gas Management System, the Reactor Ventilation System and the D_2O Recovery System.

2.1.1 Off-Gas Management System

The Off-Gas Management System treats the gases from the bleed condenser de-gassing stream, fuel transfer purge air stream, fuelling machine head-gas collection stream (Bruce 'A' and 'B' and Darlington only) and D_2O collection tank exhaust (Darlington only). The main component of this system is the charcoal delay bed that allows noble gases (especially Xe-133) to decay. Decontamination factors (DF) for Xe-133 of from 40 to 91 are the usual design values for these beds. A condenser, moisture separators and a guard bed are provided upstream of the charcoal beds to remove the moisture and organics that would adversely affect the performance of the charcoal bed. At Darlington, a recombiner is added to the bleed condenser degassing stream to oxidize all the hydrogen and deuterium in the stream. Downstream of the delay beds, the degassing stream is returned to the D_2/H_2 addition system (except Darlington) while the other streams are vented to the Active Ventilation System. A simplified schematic diagram of the Off-Gas Management System is shown in Figure 1(a).

2.1.2 Reactor Ventilation System

The Contaminated Exhaust System in the Reactor Ventilation System at Pickering 'A' and 'B', Bruce 'A' and 'B' and Darlington also provide gas treatment filters. The main functions of these systems are to remove particulates and iodine by high efficiency particulate air (HEPA) filters and high efficiency charcoal air (HECA) filters respectively. The charcoal filters are impregnated with KI for organic iodine removal. These filters are normally by-passed when no activity is measured in the air. A simplified schematic diagram of this system is shown in Figure 1(b).

2.1.3 D_2O Vapour Recovery System

The D_2O Recovery System mainly consists of molecular sieve beds to recover D_2O vapour from selected areas. Any tritium in the form of HTO or DTO is also recovered. The beds are regenerated with a hot air stream and the moisture is condensed and collected. The areas that may be connected to a D_2O Recovery System include: reactor vault and fuelling duct, moderator room, boiler room, central fuelling area and service area rehearsal facilities. A simplified schematic diagram of a D_2O Recovery System is shown in Figure 1(c). At Bruce 'B' NGS, filters designed for post-LOCA conditions to remove particulates and iodine are provided in the D_2O Recovery System.

2.1.4 Other Control Systems

Although the Tritium Removal Systems (TRS) to be installed at Pickering and Darlington do not remove tritium from the gas phase directly, they will reduce the internal radiation dose to station personnel as well as emissions of tritium to the environment. The successful operation of this system is therefore, a very important part of tritium control for normal station operation. In this system, tritium will be removed from the moderator and/or heat transport systems by vapour phase catalytic exchange and then purified by cryogenic distillation. A schematic diagram of the system is shown in Figure 2.

A dry, ambient temperature process to remove carbon-14 from moderator cover-gas was developed by Ontario Hydro Research Division. The process is simple, reliable and safe. The $Ca(OH)_2$ absorbent used in the process is inexpensive, non-toxic and thermally stable. Carbon-14 is fixed in a stable solid form ($CaCO_3$) suitable for disposal/7/. A schematic diagram of the process is shown in Figure 3. With some modifications, the process can also be applied to the nitrogen annulus gas system at Pickering 'A'.

2.2 Post-LOCA Containment Clean-Up System

The Filtered Air Discharge (FAD) System at Pickering 'A' and 'B' and the Emergency Filtered Air Discharge (EFAD) System at Darlington are designed for post-accident containment clean-up. Bruce 'A' and 'B' do not have separate EFAD Systems but the D_2O Recovery Systems have HEPA and HECA filters for post-accident gas clean-up.

This system is designed to maintain containment at a suitable sub-atmospheric pressure following an earthquake or a postulated LOCA plus impaired containment envelope accident to remove particulates and iodine from the air exhausted to the environment. At Pickering, the FAD system serves both Pickering 'A' and 'B' NGS. It consists of two parallel units of demister, air heater, pre-filter, HEPA filter, HECA filter and HEPA filter. Each filter unit is capable of handling 3400 m^3/h gas flow. The design of the EFAD system at Darlington NGS is similar to the FAD system at Pickering with two parallel filter units and a flow capacity of 5950 m^3/h for each filter unit. These systems are designed to remove:

(i) 99% by weight of the entrained moisture in an airstream containing 1.55 to 2.07 g per m^3 of entrained water and at least 99% by count, of the 2.5 to 10 μm diameter droplets.

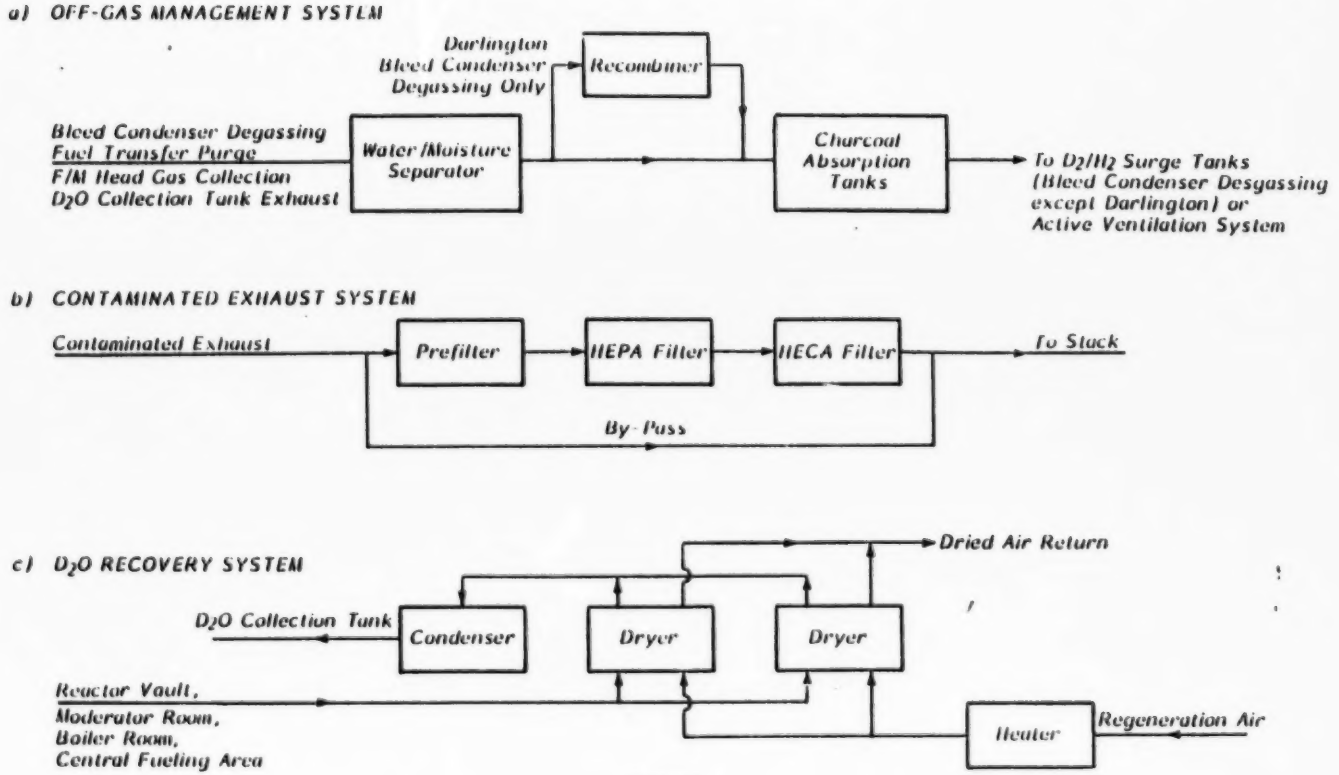

FIGURE 1

SCHEMATIC DIAGRAMS OF GAS TREATMENT SYSTEM IN NGS

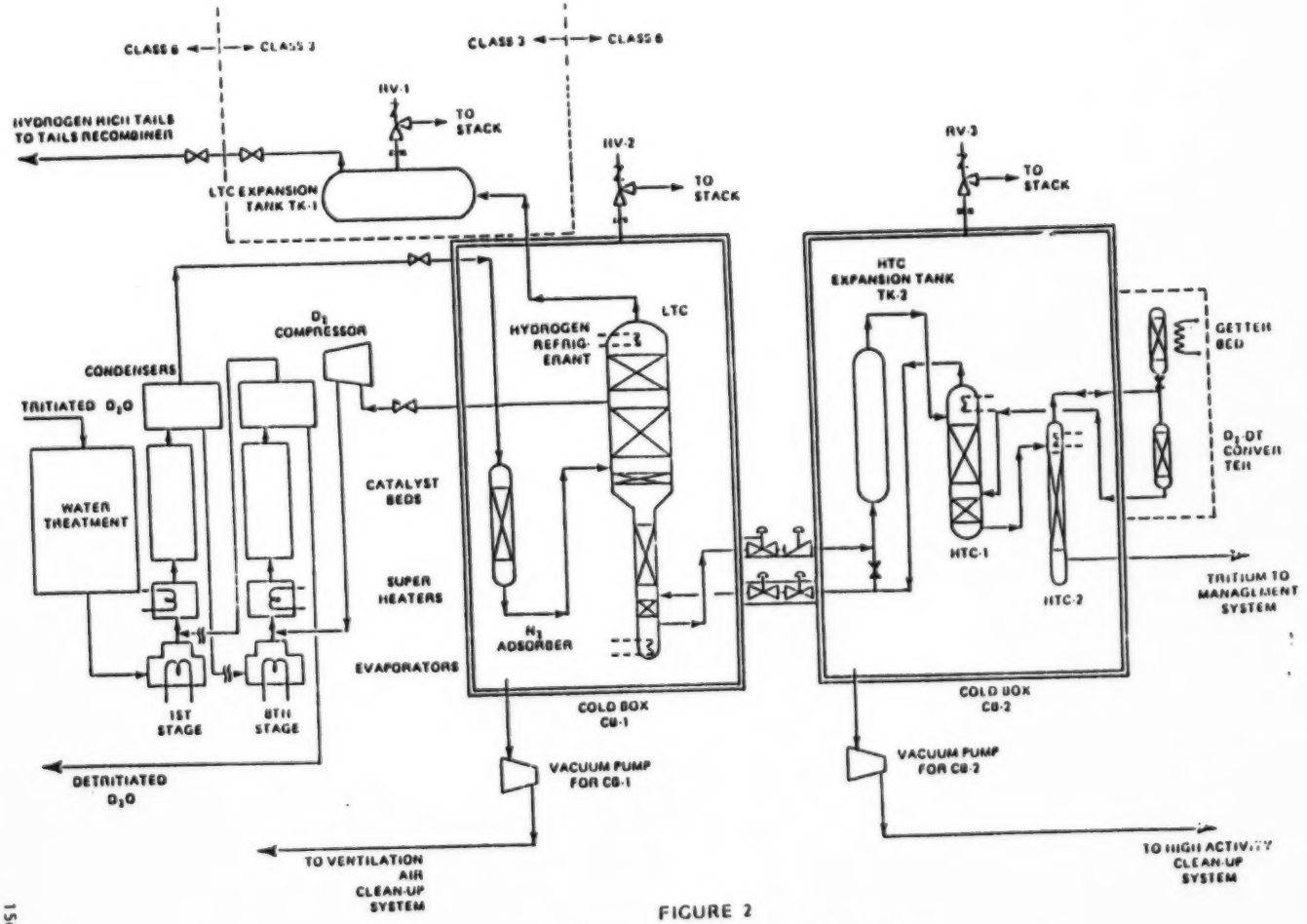

FIGURE 2

TRITIUM REMOVAL PROCESS SCHEMATIC /14/

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(ii) 99.97% by weight of >0.3 μm particles.

(iii) 99.8 to 99.99% I_2 and 99.8% CH_3I.

The FAD and EFAD Systems are not designed to remove tritium or noble gases. To limit the amount of radioactive noble gases released from the station, the amount of air discharged into the outside atmosphere must be controlled. Hence, these systems are designed to return part of the treated air back to the Vacuum Building. A simplified schematic diagram of the system is shown in Figure 4.

2.3 Fuel Reprocessing Off-Gas Treatment

A comprehensive review of the status of irradiated fuel processing off-gas treatment was carried out by Jackson/1/. The review covered iodine, tritium, carbon-14, krypton-85, nitrogen oxides, semi-volatiles and particulate control. A brief summary of the processes developed or being investigated and their status is given in Appendix C.

Processes for iodine (silver-loaded solid sorbent/8/), krypton (cryogenic trapping), and particulate control (HEPA filters) are relatively advanced and may be considered to be commercial. But the cost of silver used in the solid sorbent iodine removal process is high and complete removal of H_2O, NO_x, O_2, CO_2 is necessary to avoid freezing or an explosion hazard in the cryogenic process to trap Kr-85/9/. Therefore, further development of these processes (eg, regenerative silver sorbent) or development of alternate processes (eg, the FASTER process or solid sorbent system for controlling Kr-85) is necessary. Processes to control other radionuclides or pollutants such as voloxidation or segregation-recycle-enrichment for tritium, alkaline scrubbing or alkaline earth hydroxide sorbent for carbon-14/10/, scrubbing or catalytic reduction by H_2 or NH_3 for NO_x and denitration and sorption on iron oxide/silica for ruthenium, are all in developmental stages.

A schematic diagram of a possible off-gas treatment system is shown in Figure 5. If voloxidation is adopted to control tritium, the fuel shear and voloxidizer off-gas will be treated to remove tritium. The dissolver off-gas (DOG), after acid recovery and NO_x removal, combines with the off-gas from the tritium removal system. The combined off-gas then enters the iodine removal system, followed by the carbon-14, krypton-85 and fine particulate removal systems. Most of the carbon-14 and krypton-85 is released to this combined off-gas and very little is found in the vessel off-gas (VOG). Therefore, for the VOG, only iodine removal is required. Process modifications are being developed to release most of the iodine to the DOG so that

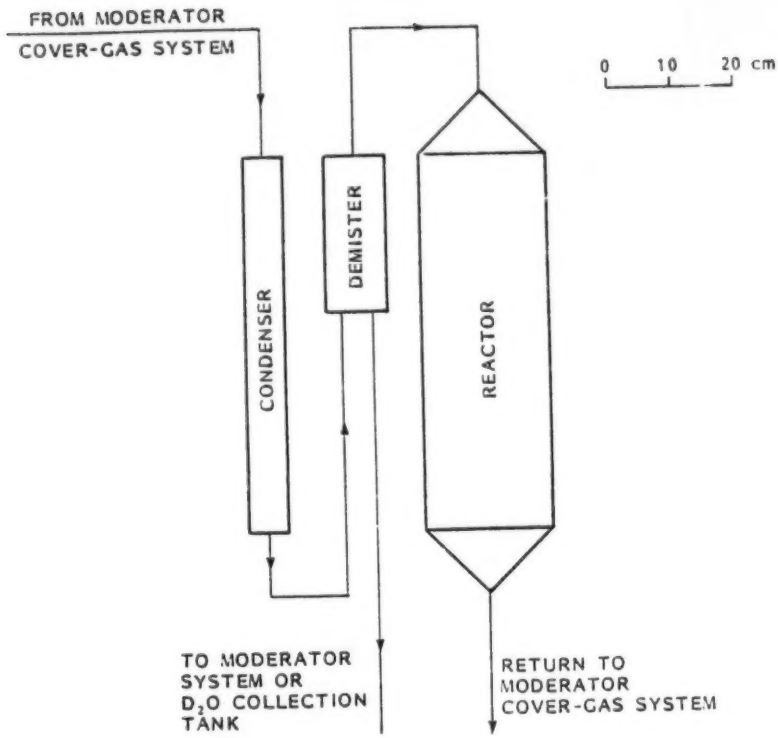

FIGURE 3

SIMPLIFIED FLOW DIAGRAM OF A CARBON-14 REMOVAL SYSTEM
FOR THE MODERATOR COVER-GAS

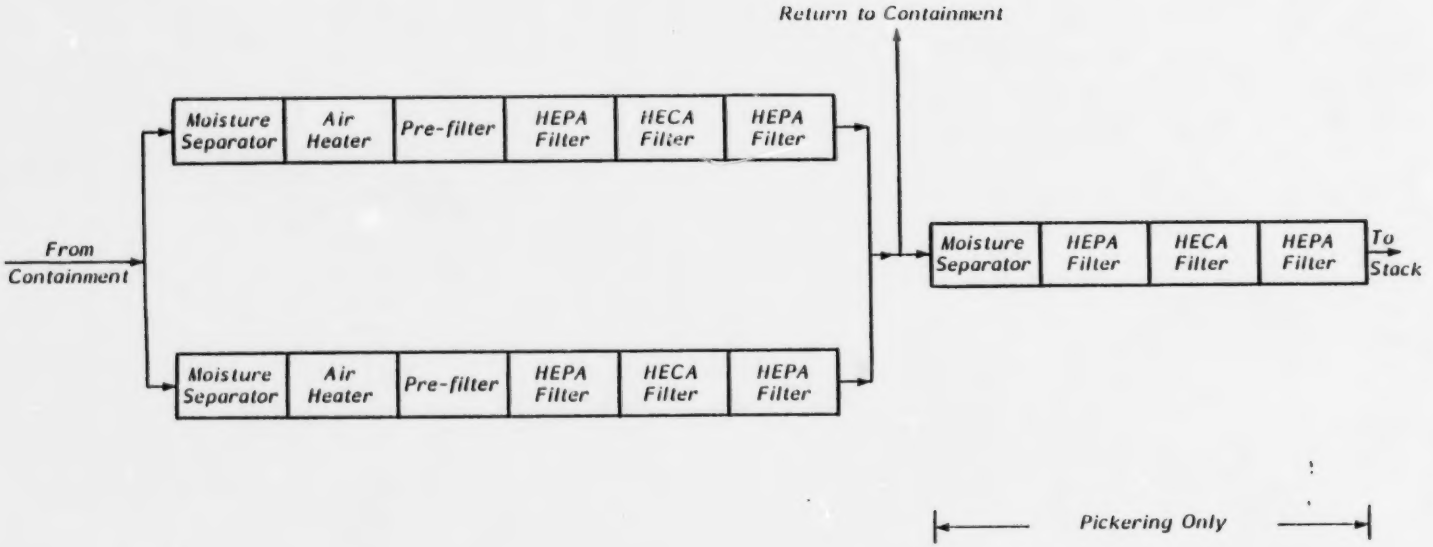

FIGURE 4

SCHEMATIC DIAGRAM OF EMERGENCY FILTERED AIR DISCHARGE SYSTEM

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iodine removal from the VOG will not be necessary/11/. Semi-volatiles, especially ruthenium, may be found in the off-gas from the high temperature high level liquid waste (HLLW) immobilization operation. Hence, a ruthenium scrubber (iron-oxide coated silica gel) is required to treat the off-gas. After ruthenium removal, this off-gas combines with the VOG for iodine and fine particulate removal.

3.0 SYSTEM COMPARISON AND EVALUATION

Gas treatment processes for normal station operation, post-LOCA containment clean-up and fuel reprocessing plants are compared in Table I for iodine, tritium, noble gases, carbon-14, semi-volatiles, particulates and NO_x and discussed in the following.

3.1 Iodine

HECA filters are being used in existing stations during normal station operation and for postulated LOCA conditions to capture iodine. However, the chemical forms and behaviour of iodine in containment during and after a postulated LOCA is not well defined. The ability of charcoal to remove iodine under post-LOCA conditions has not been proven experimentally/12/. Its application to FRP off-gas containing NO_x is not recommended because charcoal is a potential fire hazard/13/.

There are several iodine removal systems developed for FRP such as caustic scrubbing, mercuric nitrate - nitric acid scrubbing, the Iodox process and metal-doped solid sorbent systems/13/. Metal-doped solid sorbent (eg, silver impregnated zeolites, silica or alumina) systems are simple, non-combustible, the waste product is a dry solid, easily handled and contained, silver iodide is insoluble and the process is efficient for the removal of both organic and inorganic iodine species/8/. This is, therefore, the preferred process although the cost for silver is high.

3.2 Tritium

The molecular sieve dryers in the D_2O vapour recovery systems remove tritium as water vapour. The tritium recovered is recycled to the moderator. The Tritium Removal System to be installed at Pickering 'A' will remove tritium from the heat transport and moderator systems by vapour phase catalytic exchange. Tritium is then purified by cryogenic distillation and stored as metal hydride. The successful operation of the TRS is also essential to reducing in-station tritium concentration and emissions to the environment. Tritium capture in FRP has not been developed to the commercial scale. Both

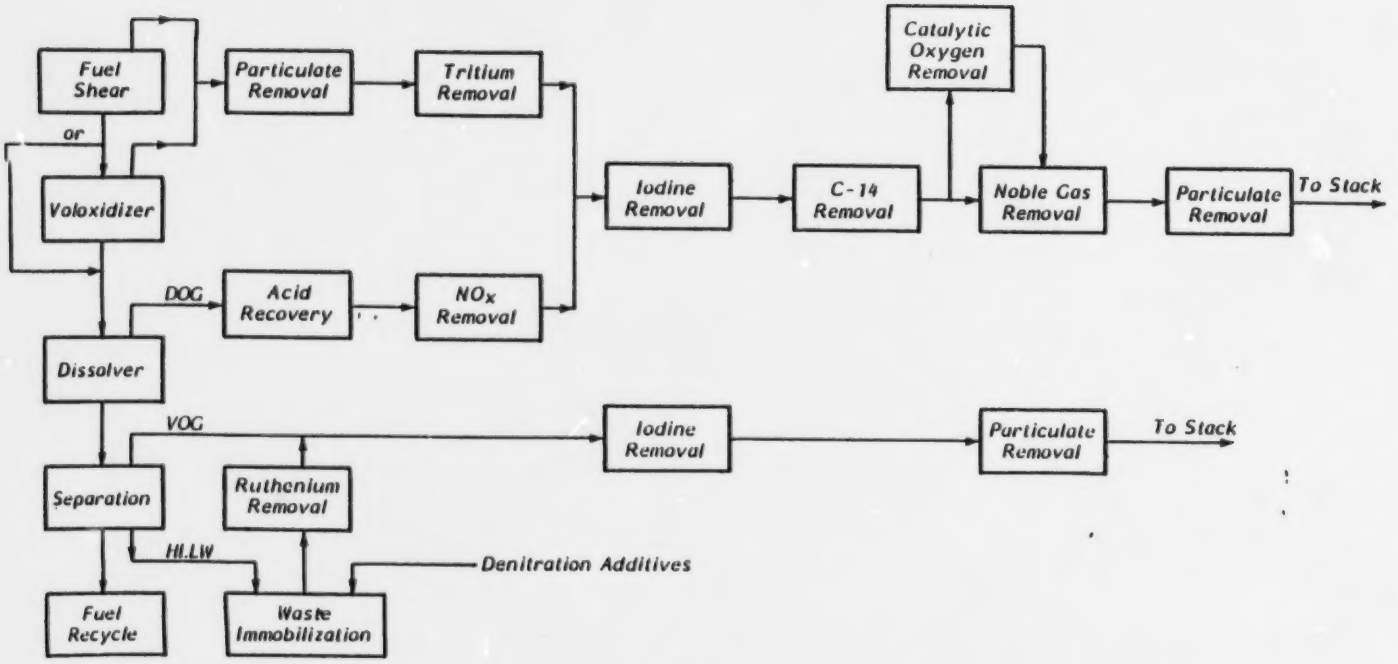

FIGURE 5
SCHEMATIC DIAGRAM OF OFF-GAS TREATMENT SYSTEMS FOR FRP

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TABLE I

COMPARISON OF GAS TREATMENT SYSTEMS AND PROCESSES

	Normal Station Operation	Containment Clean-up	Fuel Processing Plant
Iodine	charcoal filters	charcoal filters	metal-doped solid sorbent
Tritium	molecular sieve dryers Tritium Recovery System will be installed	Mist separator - molecular sieve dryer	- voloxidation - segregation-recycle - enrichment - molecular sieve dryer
Noble gases	charcoal delay beds	control may be considered	- cryogenic trapping and rectification - fluorocarbon absorption (FASTER process) - solid sorbent
Carbon-14	removal from moderator cover gas and annulus gas by $Ca(OH)_2$ may be considered	not required	- alkaline earth hydroxide absorption (wet or dry) - FASTER process
Semi-volatiles (Ru)	not required	not required	- denitration by additives (H_3PO_3, sugar, HNO_2, HCHO, HCOOH) - venturi scrubber, condenser, solvent bed & HEPA filter
Particulates	pre-filter mist separator HEPA filter	pre-filter mist separator HEPA filter	pre-filter mist separator HEPA filter
NOx	not required	not required	- acid and alkaline scrubbing - H_2 or NH_3 reduction - liquid phase additives (H_3PO_4, sugar, HNO_2, HCHO, HCOOH)

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voloxidation and segregation-recycle-enrichment processes show promise/14/ but voloxidation has not been applied to CANDU fuel and the concept of segregation and recycle has not been tested in pilot- or full-scale FRP.

3.3 Noble Gases

Charcoal beds are used in stations to delay the release of noble gases (especially Xe-133) to allow them to decay to lower radioactivity levels before release. These systems are designed to achieve a DF of 40 to 91. There are no noble gas treatment systems for post-LOCA containment clean-up. For FRP off-gas treatment, both cryogenic trapping and fluorocarbon absorption processes may be applied to capture Kr-85. The disadvantage of these two processes is that they both operate at elevated pressure. Solid sorbent systems operating at pressures close to ambient have the advantages of simplicity, safety, reliability and low operating costs, but are in an early developmental stage/16,17/.

3.4 Carbon-14

There are no gas treatment systems in stations designed for carbon-14 removal for normal or post-LOCA air clean-up. A removal system for treating moderator cover gas has been developed and a conceptual design is being prepared by Ontario Hydro. With some modification, this process will also be applicable to nitrogen annulus gas systems, the other major source of carbon-14 in the CANDU system. Carbon-14 in FRP may be captured by several processes under development including aqueous alkali systems, the FASTER process and solid sorbents, such as molecular sieves and alkaline-earth hydroxides/7,18/. The alkaline earth hydroxide system developed by Ontario Hydro may also be applicable to FRP and it offers the major advantage of direct capture and final immobilization/7/ of carbon-14 in one step. Further processing steps are required for the final immobilization of carbon-14 in other processes.

3.5 Semi-Volatiles

Most of the semi-volatiles exist as particulates under normal operating or post-LOCA conditions and are controlled by particulate removal devices such as HEPA filters. On the other hand, ruthenium is very volatile during HLLW evaporation, calcination and vitrification operations in a FRP. The use of denitrating additives to the HLLW to reduce the volatility of the semi-volatiles and ferric oxide and silica gel sorbents to capture ruthenium is near plant-scale operation.

3.6 Particulates

Particulate control technology is well established and has been widely applied in NGS, FRP and other nuclear facilities. Fibre filter, deep bed sand and deep bed fibre glass filters can all be used as pre-filters for coarse particles (>1 μm). HEPA filters are used for fine particles (with a removal efficiency of 99.97% for 0.3 μm particles).

4.0 R&D AREAS

Systems required to control the gaseous emissions of most of the radionuclides from stations to below the target 1% derived emission limit during normal station operation and systems required for post-LOCA containment clean-up are commerically available and are installed in existing stations. The main emphasis of R&D should, therefore, be on improving the operation and efficiency of these systems and developing other means that will further reduce emissions of air-borne radionuclides. To further improve emission control, it is necessary to identify the major sources of gaseous radionuclides, and to follow their spread in the stations and pathways of release to the environment so that the most effective means of control can be selected. The tracer gas technique can be used to study the spread of contaminants and develop containment schemes as discussed in an Ontario Hydro Research Division Report/20/ and should be further developed. Studies on additional control systems such as the TRS to be installed at Pickering and Darlington, the carbon-14 removal system for the moderator cover-gas and nitrogen annulus-gas and noble gas control containment clean-up, are also required.

Off-gas treatment systems for FRP, on the other hand, are mostly in a developmental stage except for iodine, noble gases and particulate control. Promising processes are being developed for tritium, carbon-14, semi-volatiles, and NO_x removal, and some have reached full-scale testing. Most of these processes have not been tested in an integrated off-gas treatment system and their performance when treating a gas with potentially interfering species is the main concern. Efforts to develop promising processes for individual radionuclides and design and testing of an integrated off-gas system are required.

A brief list of potential R&D areas for each radionuclide or pollutant is presented below. This list is not meant to be comprehensive but more of an indication of where R&D efforts should be emphasized. No priorities have been assigned to these areas.

4.1 Iodine

(i) The possible desorption of iodine from charcoal beds is an area of concern and the chemical forms and behaviour of iodine in containment during and after a postulated LOCA have not been well-defined, hence, studies on these areas are required.

(ii) For FRP off-gas treatment, the development of a regenerative silver sorbent system will reduce the cost of the process. In addition, studies of ways to remove interfering species, eg, alkanes and tri n-butyl phosphate in the VOG, will extend the use of silver sorbents to treat VOG, and therefore should be emphasized.

4.2 Tritium

(i) Further development of the technology for tritium containment and ventilation control for both stations and FRPs will reduce in-plant tritium concentrations and emissions to the environment.

(ii) Improvements to current isotopic enrichment and tritium immobilization processes will be applicable to both the TRS and FRPs.

(iii) The concept of segregation-and-recycle must be studied on a pilot scale to assess the degree of tritium transfer to the organic phase, and the effect of build-up of the various species in the FRP.

(iv) The applicability of voloxidation to irradiated CANDU fuel will have to be evaluated by experimental testing.

4.3 Noble Gases

(i) Noble gas capture may be required for post-LOCA containment clean-up in the future and control options should be evaluated.

(ii) The solid sorbent systems should be further developed because these systems operate at ambient pressures and are capable of high decontamination factors.

4.4 Carbon-14

(i) The carbon-14 removal system developed for the moderator cover gas system by Ontario Hydro should be modified and tested so that it can be applied to the nitrogen annulus gas system.

(ii) The alkaline earth hydroxide process developed for the moderator cover-gas may also be applicable to the FRP. Its development should, therefore, be extended to assess the effect of potential interfering species such as NO_x on the performance of the sorbent.

4.5 Semi-Volatiles

The control of semi-volatiles in off-gas of HLLW immobilization operations is essentially ruthenium control. Addition of denitration reagent to HLLW and capture of ruthenium with ferric oxide and silica gel sorbents should be further developed to apply to the immobilization of HLLW from CANDU fuel reprocessing.

4.6 Particulates

The design of pre-filters to improve this capability to protect HEPA filters and means to reduce the volume of the used filters are the primary R&D areas.

5.0 CONCLUSIONS AND RECOMMENDATIONS

Several gas treatment systems are installed in stations to control emission of radionuclides during normal station operation. These include Off-gas Management System (for noble gas retention and particulate removal), Active Exhaust System (for iodine and particulate control), and D_2O Vapour Recovery System (for recovery of deuterium and tritium as water vapour). The FAD or EFAD Systems are designed to control emissions of iodine and particulate during post-LOCA containment clean-up. These systems are commercially available. The emphasis of R&D on gas treatment systems for stations should be on the following areas:

1. Improvement of the operation and efficiency of the existing systems.

2. Development of the tracer gas technique to identify the release pathways of air-borne radionuclides and to evaluate the effectiveness of any control scheme.

3. Research in support of the TRS to ensure the success of its operations.

4. Modification of the carbon-14 removal system developed by Ontario Hydro for nitrogen annulus gas.

5. Studies on the desorption of iodine from charcoal beds and the chemical forms and behaviour of iodine in containment during and after a LOCA.

6. Selection of a noble gas removal system for post-LOCA containment clean-up.

Most of the off-gas treatment processes for FRP have not reached commercial stage. Some of the first generation processes have many disadvantages and the development of alternative processes may be necessary. Recommended research areas include:

1. Development of control processes for individual radionuclides or pollutants especially tritium, noble gas, carbon-14 and NO_x.

2. Design and testing of an integrated off-gas treatment for FRP.

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Reviewed by:

J.A. Chadha
Area Coordinator
AECL Technical Assistance
Program

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APPENDIX A

GAS TREATMENT SYSTEMS FOR NORMAL STATION OPERATION

Major gas treatment systems in a station under normal operating conditions include the Off-gas Management System, the Ventilation System and the D_2O Recovery System. The design of these systems at Pickering 'A' and 'B', Bruce 'A' and 'B', and Darlington will be discussed and compared in this Appendix. A Tritium Removal System will be retrofitted at Pickering 'A' to reduce in-station tritium concentrations and tritium emission although it is not a gas treatment system.

A.1 Off-Gas Management System

The Off-gas Management System in a station is designed to reduce the release of the fission product radioactive noble gases from the station. The system also recovers deuterium and heavy water in the process.

The Off-gas Management Systems are slightly different for different stations. At Pickering 'A' and 'B' and Bruce 'A' and 'B', there are two trains of charcoal beds treating bleed condenser degassing stream and fuel transfer purge (and F/M head gas collection strain at Bruce 'A' and 'B') separately. At Darlington, a recombiner is added to the bleed condenser de-gassing and D_2O collection tank exhaust streams, and a common charcoal delay bed system treats all the off-gas streams. Schematic diagrams illustrating these systems are shown in Figure A.1.

A.2 Active Ventilation System

Some of the ventilation systems in stations are equipped with iodine and particulate filters to treat the contaminated exhaust. These filters are associated with the Reactor Building Ventilation System in all stations. They are normally by-passed except when high iodine, particulate or gamma activity levels in the exhaust duct are detected. The capacity of the filter systems and filter components provided at each station are summarized in Table A.2.

A.3 D_2O Recovery System

The purposes of the D_2O Vapour Recovery Systems are:

(i) to recover D_2O leakage within the dried areas and to reduce D_2O losses due to exhaust or vapour escape from these areas;

(ii) to reduce tritium or fission products and activated particle contamination inside the dried areas or in adjacent accessible areas due to leakage or vapour migration;

(iii) to clean-up dried areas after minor accidental system failure, temporary high leak rates, or prior to shut down or routine maintenance and;

(iv) to minimize downgrading caused by air or light water leakage into the dried areas.

The systems at Bruce 'B' also serves to maintain containment at a sub-atmospheric pressure following an earthquake and to provide a means for controlled discharge exhaust during long term post-LOCA operation and to prevent containment repressurization.

The reactor building areas which are subject to D_2O leakage are classified into several separate systems containing atmospheres of different D_2O and tritium isotopic content, and each area is provided with separate D_2O recovery and collection equipment to prevent unnecessary downgrading. The areas and D_2O vapour recovery system capacity in these areas for various NGS are summarized in Table A.3.

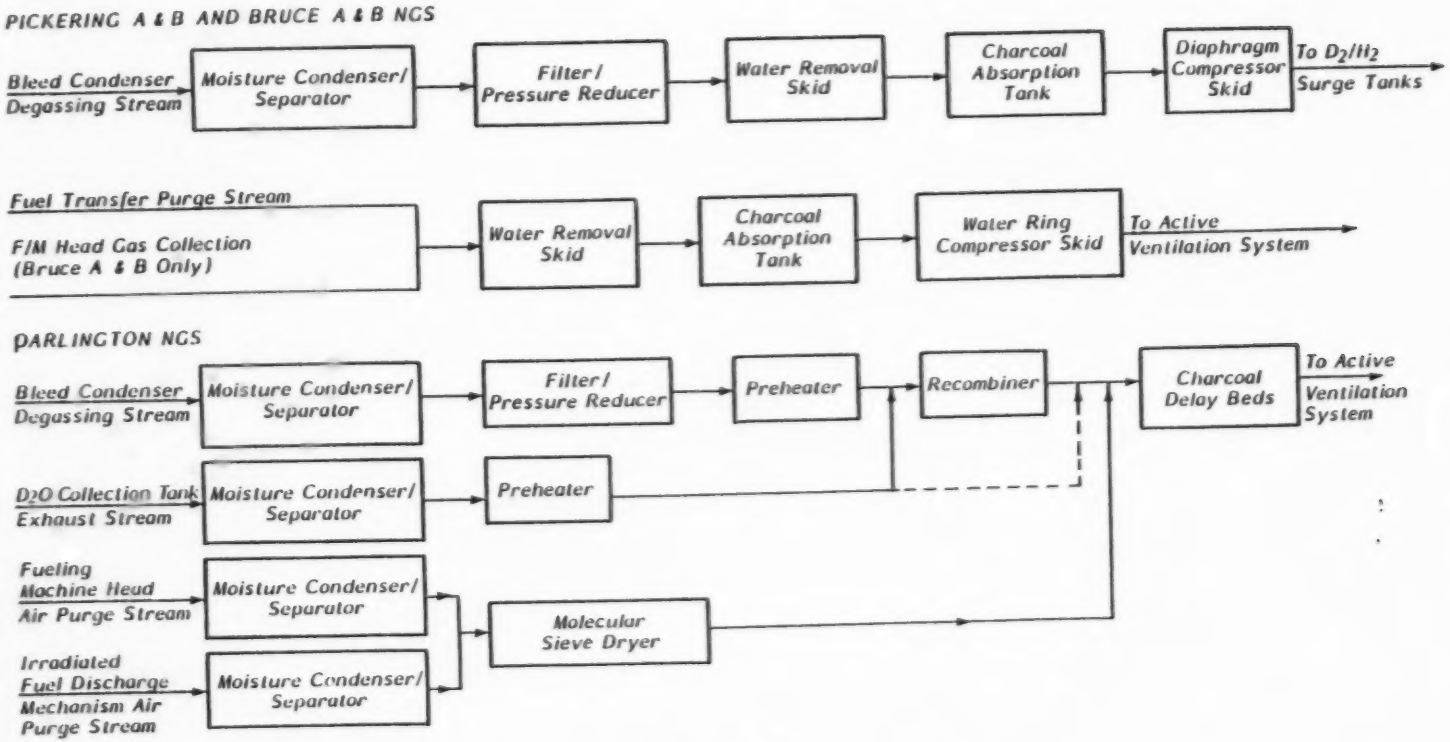

FIGURE A-1

SCHEMATIC DIAGRAMS OF OFF-GAS MANAGEMENT SYSTEMS IN NGS

TABLE A.I

SUMMARY OF OFF-GAS MANAGEMENT SYSTEM DESIGN

Station	Off-Gas Streams	Flow Rate (m^3/h)	DF for Xe-133	Gas After Treatment
Pickering 'A' (DM-44-79320/67932)	Bleed Condenser Degassing	3.4 (0.34/unit)	40	to D_2/H_2 addition system
	Fuel Transfer Purge	1.7-13.6	40	to the active ventilation system
Pickering 'B' (NK30-79320)	Bleed Condenser Degassing	3.4	40	to D_2/H_2 addition system
	Fuel Transfer Purge	13.6	40	to irradiated fuel bay ventilation system
Bruce 'A' (21-79320/67932)	Bleed Condenser Degassing	3.4 (0.85/unit)	40	to D_2/H_2 addition system
	Fuel Transfer Purge and F/M Head Gas Collection	17-68 (3.4 avg) (2.4 avg)	40	to active exhaust system
Bruce 'B' (DD-29-79320/67932)	Bleed Condenser Degassing (70%)	3.4 (0.85/unit)	40	to D_2/H_2 addition system
	Fuel Transfer Purge (15%)	17 max (4.76 ave)	40	to active exhaust ventilation
	F/M Head Gas Collection (15%)	27.2 max (3.4 ave)	40	to active exhaust ventilation
Darlington (38-79800)	Bleed Condenser Degassing	8.5/unit	91	to recombiner; to dryer and charcoal bed then active ventilation system
	D_2O Collection Tank Exhaust	3.4/unit	91	
	Fuelling Machine Head Air Purge	27.2 max (3.4 ave)	91	
	Fuel Discharge Air Purge	34 max (4.1 ave)	91	

*(System Classification Index (SCI) numbers)

TABLE A.II

SUMMARY OF DESIGN INFORMATION OF CONTAMINATED EXHAUST SYSTEMS

Station	Capacity (m^3/h)	Major Components
Pickering 'A' (44-73130)	11 050	HEPA filter, charcoal filter
Pickering 'B' (30-73130)	13 200	demister, heater, prefilter, HEPA filter, HECA filter
Bruce 'A' (21-73220)	102 000	prefilter, HEPA filter and HECA filter
Bruce 'B' (29-73220)	129 000	prefilter, HEPA filter and HECA filter
Darlington (38-73220)	153 000	prefilter, HEPA filter and HECA filter

(SCI numbers)

TABLE A.III

SUMMARY OF CAPACITIES OF D_2O VAPOUR RECOVERY SYSTEMS

Station	Areas with D_2O Recovery System	Capacity (m^3/h)
Pickering 'A' (44-72200)	Boiler Room	6 800
	Moderator Room	6 800
	Reactor Vault	6 800
Pickering 'B' (30-72200)	Boiler Room	
	Reactor Vault or Fuelling Machine Vault	4 250
	Fuelling Machine Service Rooms and Auxiliary Room	4 250
	Moderator Room	
Bruce 'A' (21-38300)	Reactor Vault and Fuelling Duct	4x6 800
	Central Fuelling Area	2x3 400
	Service Area Rehearsal Facilities	850
	Moderator HX and Pump	possible future dried area
Bruce 'B' (29-38300) (29-38300/63831)	Reactor Vault and Fuelling Duct	4x6 800
	Central Fuelling Area	
	Service Area Rehearsal Facilities	
	Confinement	
Darlington (38-38300/38340)	Reactor Vault and Fuelling Duct	5x6 800
	Fuel Handling Service Area and Rehearsal Facilities	13 600
	Moderator HV and Pump	

(SCI numbers)

APPENDIX B

POST-ACCIDENT CONTAINMENT GAS CLEAN-UP SYSTEMS

Following an accident, a gas treatment system will operate to maintain the negative pressure of the containment. This system provides a controlled release path which filters radioactivity (iodine, particulates and tritium as entrained moisture) from the air exhausted to the environment. The gas treatment system is different for different stations. Pickering 'A' and 'B' have a common containment gas clean-up system called Filtered Air Discharge (FAD) System. Bruce 'A' and 'B' do not have a separate system for containment gas clean-up, but at Bruce 'B', the D_2O Vapour Recovery System has filters designed for post-LOCA conditions to remove particulate and iodine. Darlington will have an Emergency Air Discharge System for containment clean-up. These systems are compared in Table B.1.

The filter systems in the various stations consist of the same components although the sizing of the components may be different. The components are: moisture separator, heater, pre-filter, HEPA filters and charcoal absorbers.

TABLE B.I

SUMMARY OF DESIGN INFORMATION ON POST-LOCA CONTAINMENT CLEAN-UP SYSTEM

Station	Post-Accident Gas Treatment System	Capacity (m^3/h)	Design Requirement
Pickering 'A' and 'B'	Filter Air Discharge System	1 700	- maintain containment sub-atmospheric - iodine removal efficiency: organic 99.0% elemental 99.5% - particulate removal efficiency: 99.97% for >0.3μm - demister efficiency: 99% for 1-10μm droplets
Bruce 'A'	Do not have separate gas treatment system	-	-
Bruce 'B'	System associated with D_2O vapour recovery unit	850	- iodine removal efficiency: 99.95% for I-131, HOI and CH_3I - particulate removal efficiency: 99.97% for >0.3 μm - demister efficiency: 99% for 1-10 μm droplets
Darlington	Emergency Filtered Air	3 400	- maintain containment at 0.13 kPa below atmospheric - iodine removal efficiency: 99.8% for I_2 and CH_3I - particulate removal efficiency: 99.97% for 24-32 gr/m^3 air

APPENDIX C

GAS TREATMENT SYSTEMS FOR FUEL REPROCESSING PLANT

A comprehensive review of the gas treatment systems for FRP was carried out by Jackson/1/. Processes for controlling emissions of iodine, tritium, carbon-14, krypton-85, NO_x, semi-volatiles and particulates were reviewed. Treatment processes for each of the radionuclides and pollutants and their status of development are summarized in Table C.1 and discussed briefly in the following.

C.1 Iodine

Several iodine removal processes have reached commercial scale or near commercial scale. These include caustic scrubbing, activated carbon sorbents, mercuric nitrate-nitric acid scrubbing, the Iodox process and metal-doped solid sorbents. Metal-doped solid sorbents (eg, silver-impregnated zeolites, solica or alumina) systems have many advantages over other processes/8/ such as:

(i) the system is simple;

(ii) the adsorbent is not flammable and hence not a fire hazard;

(iii) the final product is a dry solid and the chemisorbed iodine is insoluble and can be easily handled and disposed;

and

(iv) the process is efficient for the removal of both organic and inorganic iodine species.

Metal-doped solid sorbent systems are, therefore, the preferred system to capture iodine. The main drawbacks of this process are the high cost of silver and the fact that it is less effective for VOG due to the interference of the solvent from the VOG. Regenerative transfer systems using Ag°Z and PbX zeolites are being developed to minimize the use of silver. Process modifications are also being developed to release most of the iodine to the DOG so that iodine removal from VOG is no longer necessary/13/. The alternative is to selectively remove the interfering species such as alkanes and TBP from the VOG prior to iodine removal.

C.2 Tritium/14/

Tritium control has not reached commercial scale implementation. Head end isolation by voloxidation and segregation-

recycle operation following by isotopic enrichment are the two approaches that may be adopted. In the voloxidation process, tritium release efficiencies from the fuel are high and tritium is concentrated in a small gas stream. But the problems of component maintenance, process control, and dust and criticality control may be difficult to solve. Segregation and recycle of process streams within the Purex plant following by isotopic enrichment may be effective in tritium control. This scheme provides a very attractive alternative to voloxidation but is only a concept at this stage. In particular, the effect of build-up of the various species on the process and loss of tritium to the organic phase is not known. Isotopic enrichment techniques also require further development for separating tritium from HTO/H_2O.

C.3 Carbon-14/7,18/

Carbon-14 (as $^{14}CO_2$) capturing processes can be divided into two categories: liquid and solid sorbents. Liquid sorbents include fluorocarbons (FASTER process) and aqueous alkalis (Na or Ca). Solid sorbents used for carbon-14 removal include molecular sieves and alkali earth (Ba and Ca) hydroxides. The use of solid sorbents simplifies CO_2 removal because the complexity of pumping scrubber liquor, controlling feed composition, solid-liquid separation and contaminated water treatment are avoided. However, molecular sieve sorbents require regeneration which creates a concentrated gaseous stream to be treated by liquid sorption processes. The solid alkaline earth hydroxide processes are the only processes that avoid liquid sorption completely and offer the advantage of direct capture are therefore, preferred. The process developed by Ontario Hydro for moderator cover-gas may also be applicable for FRP; however, the effect of potentially interfering species such as NO_x must be studied.

C.4 Krypton-85

Both cryogenic trapping and the fluorocarbon (FASTER) processes have been successfully developed to separate krypton from FRP off-gas streams. The cryogenic process is closer to commercial application but is more complex since it involves several subsystems for removal of interfering species such as CO_2, NO_x and O_2/9/. The FASTER process can tolerate these components and is, therefore, substantially simpler but the final separation of krypton from the fluorocarbon sorbent has not been developed yet. Both of these first generation processes operate under pressure, on the other hand, solid sorbent systems that are still at an early stage of development, operate at close to ambient pressure and offer the advantages of simplicity, safety, reliability and low operating costs/16,17/. These systems may be superior to both of the first-generation processes and should be developed further.

TABLE C.I

STATUS OF GAS TREATMENT PROCESSES FOR FRP

Radionuclides	Separation Process	DF* Achievable	Status	Comments
Iodine (I-129)	Metal-Doped Solid Sorbent	10^2 - 10^6	pilot scale at Karlsruhe field testing at Marcoule	Low efficiencies for organic iodides. High cost of silver.
	Mercurex	10^2	bench scale	Process complex with liquid waste
	Iodox	10^4	pilot scale	Process complex with liquid waste costly metal required for construction. NOx removal required.
	Alkaline Scrubbing	2 - 10	commercially available	No organic I_2 removal, liquid waste
	Carbon Sorbents	10^3 - 10^4	commercially available for NGS ventilation	Fire hazard
Tritium	Segregation, Recycle & Enrichment	5 - 20	conceptual, enrichment-bench scale	Long term effect of recycling not known.
	Recycle, Segregation	10^3	conceptual, applied to D_2O/DTO at CECE	Not demonstrated for H_2O/HTO, occupational exposure from HTO high.
	Voloxidation	10^2	bench scale	
Carbon-14	Alkaline-earth Hydroxide	10^2 - 10^3	bench scale	Process simple, interference from NOx, I_2 not known.
	Aqueous Alkali	10^2 - 10^3	pilot scale	Process complex with liquid waste
	Molecular Sieve	10^2	pilot scale	Commercially available; final immobilization not developed.
	FASTER	10^2	pilot scale	Stream contains Kr, NOx, I_2 and CO_2; further separation and fixation not developed
Krypton-85	Cryogenic	10^2 - 10^3	commercially available	Removal of I_2, H_2O, CO_2, O_2, NOx required
	FASTER	$>10^2$	pilot scale	Can tolerate contaminants
	Solid Sorbent	10^2 - 10^3	bench-pilot scale	Removal of contaminants necessary
	Selective Membrane		bench scale	High pressure may be necessary
Semi-volatiles (Ru, Te)	Denitrating with Formaldehyde and Formic Acid	-	pilot scale	Very promising, further study of the chemistry and large scale testing required
	Ferric Oxide, Manganese Oxide, Silica Gel Adsorber			Tests in France successful
NOx	Aqueous Scrubbing	-	commercially available	Traces of NOx remain in scrubbed gas
	Catalytic Reduction With H_2 or NH_3	-	bench scale	Further bench scale study necessary
Particulate	Pre-filter	2 - 3	commercially available	Volume reduction of spent filter required
	Demister	10^2	commercially available	Volume reduction of spent filter required
	HEPA Filter	10^3	commercially available	Volume reduction of spent filter required

* For DF required for each radionuclide, see reference 19

C.5 Nitrogen Oxides (NO_x)

The removal of NO_x is required to conserve the acid for plant recycle, to minimize its interference with other radionuclide removal systems, and to reduce the volatility of ruthenium. A dilute nitric acid scrubbing system or a simple condenser to lower the humidity and temperature is commonly used for acid recovery and recycle alakli scrubbing is also used for NO_x removal. The balance of NO_x in the DOG may be removed by a catalytic gaseous reduction system in which either hydrogen (non-selective) or ammonia (selective) may be used as the reductant. To reduce the volatilization of ruthenium in the high level liquid waste (HLLW) during immobilization operation, organic reductants such as formic acid and formaldehyde may be added to the HLLW to destroy the nitrate species with the generation of harmless N_2O or N_2 and reduction of ruthenium volatility. Other additives such as phosphorous acid, sugar and nitrous acid were also found to be effective.

C.6 Ruthenium and Semi-volatiles

Most of the semi-volatile species except ruthenium can be collected by cooling followed by cyclones and HEPA filter. Ruthenium is the semi-volatile that is the most difficult to control. The volatility of ruthenium may be reduced by the use of denitrating additives such as formaldehyde and formic acid to the HLLW before immobilization. Ruthenium is captured from off-gas of the immobilization operation by passing the off-gas through a condenser, ferric oxide and silica gel absorbers and HEPA filters.

C.7 Particulate Control

Pre-filters for coarse particulate control, mist eliminators for liquid droplet control and HEPA filter for fine particulate collection are well developed and commercial products are well-demonstrated in existing nuclear facilities. Information on these equipment are available for design and costing purposes.